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Measurement of the state-specific differential cross section for the $\text{H} + \text{D}_2 \rightarrow \text{HD}(v'=4, J'=3) + \text{D}$ reaction at a collision energy of 2.2 eV

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HI and D_2 are coexpanded into a vacuum chamber. The photolysis of HI at 212.8 nm initiates the $\text{H} + \text{D}_2$ reaction. The $\text{HD}(v=4, J'=3)$ velocity distribution is determined by analyzing the time-of-flight profile of HD^+ ions produced by delayed pulsed field ionization of long-lived Rydberg states. The angular distribution is deduced using the law of cosines (photoloc technique). © 1995 American Institute of Physics.

I. INTRODUCTION

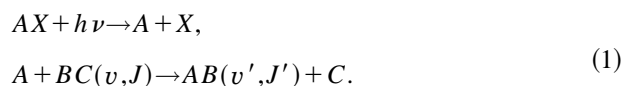
The most intuitive way of measuring the differential cross section of a bimolecular reaction is to use crossed molecular beams.¹ For the $\text{H} + \text{H}_2$ reaction and its isotopic variants, this technique is usually able to resolve product vibrational states.^{2–8} Recently, the first rovibrational-state-resolved crossed beam measurement of the $\text{H} + \text{H}_2$ reaction system was accomplished for $\text{H} + \text{D}_2(v=0, J=0)$ by Welge and co-workers,^{9–11} at collision energies of 0.53 and 1.28 eV, using the novel technique of Rydberg-atom time-of-flight spectroscopy.¹² This technique relies on the fact that D atoms associated with HD molecules in different rovibronic states carry different amounts of kinetic energy. Therefore product rovibronic state resolution can be obtained by measuring the kinetic energy of the D atoms with sufficient resolution, which is limited by space-charge effects in conventional time-of-flight (TOF) spectrometers. By preparing high Rydberg states of D atoms and ionizing them with a high-voltage field immediately prior to detection, Welge and co-workers were able to minimize space charge and resolve rovibrational states of the HD product from the TOF of D atoms at several scattering angles in the forward-scattering hemisphere. In addition to the great success of the crossed beam technique, recent years have also witnessed the evolution of alternative techniques that extract information about differential cross sections by studying photoinitiated bimolecular reactions in a single gas flow (or beam).^{13–24} The improved sensitivity of these techniques should make possible more detailed studies, such as the product-polarization dependence of the differential cross section.²⁴ These techniques include velocity selected double resonance (VSDR),¹⁶ velocity-aligned photo-fragment dynamics,¹⁷ and photoinitiated bulb reactions.²² It has been suggested in a recent paper²⁴ that this class of studies be called “photoloc” experiments, for photoinitiated bimolecular reactions where analysis is based on the law of cosines.

In this communication, we report a photoloc measurement for the state-specific differential cross section of the reaction $\text{H} + \text{D}_2 \rightarrow \text{HD}(v'=4, J'=3) + \text{D}$ at a collision energy of 2.2 eV. To date, most experiments and calculations on this fundamental reaction were performed for significantly lower collision energies. The geometric phase originating from the

Jahn–Teller effect has been shown to be important in differential as well as integral cross sections for the $\text{H} + \text{H}_2$ reaction family.^{25–31} By increasing the total energy of the reaction, we can test the importance of the geometric phase as the conical intersection of the H_3 potential energy surfaces is approached.

II. PRINCIPLES OF THE PHOTOLOC EXPERIMENT

In a typical photoloc experiment, a gas mixture of AX and BC is coexpanded into a vacuum chamber. AX is then photolyzed to initiate the reaction sequence:



Consider the ideal case where A is translationally monoenergetic, BC is in a single rovibronic state, C acts as a spectator (or is restricted to one state), and AB is detected in a specific rovibronic state. The center-of-mass (c.m.) speed u of the A, BC collision partners is fixed because AX is stationary relative to BC prior to photodissociation. The c.m. speed u_{AB} of the $\text{AB}(v', J')$ product is determined by conservation of energy. Therefore a measurement of the laboratory-frame speed v_{AB} of the $\text{AB}(v', J')$ product gives the scattering angle by applying the law of cosines. In practice, the $\text{AB}(v', J')$ laboratory velocity distribution (or a component of it) is measured, rather than the speed distribution. The relationship between the laboratory velocity distribution, $f(v_{AB})$, and the normalized differential cross section, $(1/\sigma) d\sigma/d\Omega$, is²²

$$\begin{aligned} f(\mathbf{v}_{AB}) &= \frac{1}{2v_{AB}uu_{AB}} \left(\frac{1}{\sigma} \frac{d\sigma}{d\Omega} \right) \\ &\times [1 + \beta_{\text{react}}(\mathbf{v}_{AB}) P_2(\hat{\mathbf{v}}_{AB} \cdot \hat{\mathbf{e}})], \end{aligned} \quad (2)$$

where $\hat{\mathbf{e}}$ is the direction of the electric vector of the photolysis laser, $\beta_{\text{react}}(\mathbf{v}_{AB}) = \beta_{\text{photo}} P_2(\hat{\mathbf{v}}_{AB} \cdot \hat{\mathbf{u}})$, P_2 is a second-order Legendre polynomial, and β_{photo} is the anisotropy of the A photofragment following photolysis of AX.

In our experiment, a component of the velocity distribution, $f_{AB}(v_x \approx 0, v_y \approx 0, v_z)$, is measured. The laboratory reference frame is chosen to be the Cartesian coordinate system where the x axis is in the probe laser propagation direction,

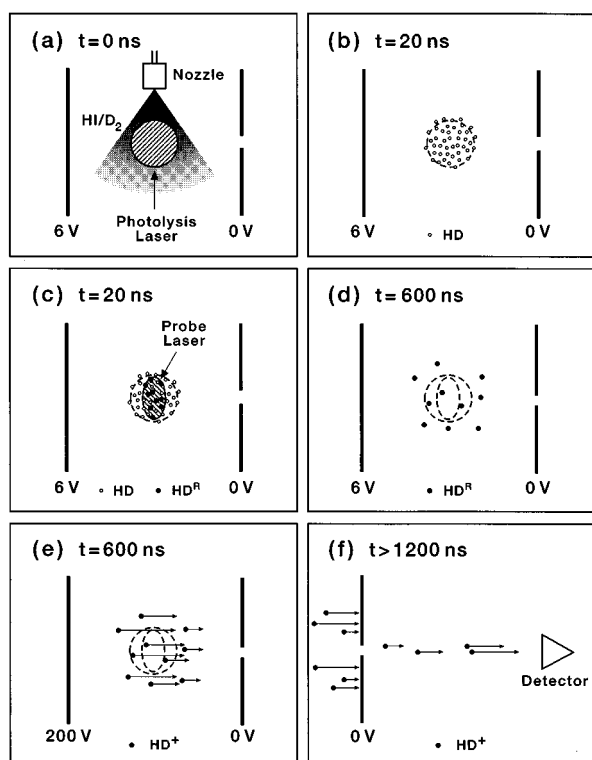


FIG. 1. Schematic diagram for the measurement of the state-specific differential cross section of the $\text{H}+\text{D}_2\rightarrow\text{HD}$ ($v'=4, J'=3$)+D reaction. (a) The reaction is initiated in a pulsed expansion of HI/D_2 by photolysis of HI at 212.8 nm. (b) The reaction proceeds for 20 ns to form products. (c) Probe lasers are fired along the x -axis (into the page) to excite nascent HD ($v'=4, J'=3$) product to high Rydberg states HD^R ; the VUV probe laser is tuned to the center of the HD Doppler profile to select $v_x\approx 0$. (d) Rydberg HD molecules drift for 600 ns. (e) A high voltage pulse is applied to field ionize HD Rydberg molecules and eject the resulting ions into the TOF tube. (f) A slit in the TOF tube selects HD ions with near-zero velocity projections along the y -axis ($v_y\approx 0$). The velocity component v_z is measured by time of flight. The beam sizes of the photolysis and probe lasers are indicated by a circle and an ellipse, respectively.

the y axis is defined by a slit in the TOF, and the flight axis forms the z axis. The relationship between $f_{AB}(v_x\approx 0, v_y\approx 0, v_z)$ and the differential cross section is not so straightforward as that shown in Eq. (2), and details of the analysis will appear in a subsequent paper.³²

III. Experiment

The apparatus is similar to that described previously.²³ A gas mixture of 5% HI (Matheson, 98%) in D_2 (Cambridge Isotope, 99.8% D) is introduced into the reaction chamber through a 400- μs pulsed valve (General Valve 9-442-900, 0.8-mm orifice). The D_2 molecules are assumed to be unaligned. The radiation of the fifth harmonic (212.8 nm, 2.5 mJ) of a Nd:YAG laser (Spectra Physics, GCR-5) is used to photolyze HI [Fig. 1(a)] and produce “hot” H atoms. These H atoms are allowed to react with D_2 molecules for 20 ns [Fig. 1(b)]. Only the fast H atoms that correlate with the production of ground-state $I(^2P_{3/2})$ atoms can produce HD in the rovibronic state we detect. The radiation from a narrow bandwidth (0.5 cm^{-1}) vacuum ultraviolet (VUV) laser (130.60 nm, 200 nJ) excites HD from the ground state

($X\ ^1\Sigma_u^+ v'=4, J'=3$) to the first excited state ($B\ ^1\Sigma_u^+ v=0, J=2$). The VUV bandwidth is narrower than the Doppler width of the transition, so only HD molecules with near-zero velocity projections along the x -axis ($v_x\approx 0$) are excited. The radiation from a Nd:YAG (Spectra Physics, GCR-2) pumped dye laser (Lambda Physik, LPD3000) is frequency doubled ($\sim 292.5\text{ nm}$, 12 mJ) and used to excite HD ($B\ ^1\Sigma_u^+ v=0, J=2$) to a high-lying *gerade* Rydberg state [Fig. 1(c)]. This Rydberg state is a complex resonance³³ between the “22d3” and high members ($n\approx 106$) of the $N^+=1$ series, which both converge to the $v^+=0$ state of HD^+ . The Rydberg-tagged HD molecules are allowed to drift for about 600 ns [Fig. 1(d)]. A 70 V/cm electric field is then switched on to ionize HD Rydberg molecules and eject the ions into the TOF tube [Fig. 1(e)]. A 16 mm \times 2 mm slit inside the TOF tube selects ions with near-zero velocity projections along the y -axis ($v_y\approx 0$). The TOF spectrum of HD^+ ions is recorded [Fig. 1(f)] from which the HD velocity distribution $f_{\text{HD}}(v_x\approx 0, v_y\approx 0, v_z)$ is determined.

Two major difficulties had to be overcome. First, HI, I (from HI photolysis) and I_2 (from HI decomposition) in the gas mixture can be ionized to release photoelectrons. These photoelectrons, when accelerated by the extraction pulse, can ionize D_2 in an avalanche process that leads to distortions in TOF profiles. Therefore a 6 V bias is applied to the extraction plate to remove photoelectrons before the extraction pulse. Gas flow is also restricted to reduce background ions. Second, product HD molecules with the highest velocities (corresponding to forward-scattered products) tend to escape from the photoexcitation region faster than those with the lowest velocities (corresponding to backward-scattered products), which causes a preferential loss of forward scattering signal in the differential cross section. To avoid this bias from fly out, the time delay between photolysis and probe lasers must be less than 20 ns.

The lifetime of HD Rydberg molecules is measured to be on the order of microseconds under our experimental conditions; therefore, no lifetime correction is needed. The lifetime measurement of HD *gerade* Rydberg states, along with spectroscopic and dynamical studies of these states, will be presented in another paper.³³ A small difference (less than 10%) is observed when the polarization of the VUV probe laser is rotated 90°. This effect suggests that the differential cross section depends weakly on product HD rotational alignment—a topic that warrants further study.²⁴

IV. DATA AND ANALYSIS

The thin line in Fig. 2 shows a typical TOF spectrum. The first peak corresponds to HD Rydberg molecules moving initially away from the detector (backward traveling) and the following peaks correspond to those moving initially toward the detector (forward traveling). Backward-traveling molecules move at first toward higher potential regions than forward-traveling ones. When they are ionized by the extraction pulse, they reach higher velocities [shown in Fig. 1(e) as longer arrows] and arrive at the detector earlier than forward-traveling molecules, despite the longer distances they traverse. In principle, either the backward- or the forward-

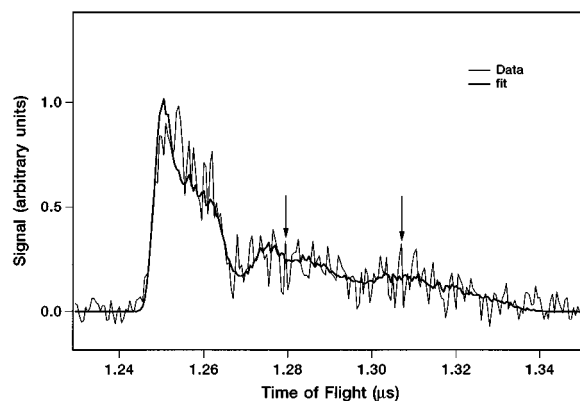


FIG. 2. Experimental TOF profile (thin line) and Monte Carlo simulation (thick line) of the HD ($v'=4, J'=3$) reaction product. The arrows point to features that are caused by structures in the differential cross section. The backward-traveling ions appear at earlier times ($\leq 1.265 \mu\text{s}$) than the forward-traveling ions.

traveling peak can be used to obtain the HD velocity distribution. In this experiment, however, the forward-traveling peak is better resolved.

To determine a differential cross section from the data presented in Fig. 2, deviations from the ideal photoloc experiment must be considered. In the ideal case, the translational and rotational temperatures of the HI/D₂ mixture approach 0 K and the speed of the HD product is measured with near-infinite resolution. If these conditions are met, the differential cross section can be determined directly using Eq. (2). To interpret the data reported here, however, we must consider the finite translational temperature of the pulsed expansion (~ 50 K), the finite rotational temperature of the HI precursor (~ 50 K), the finite rotational temperature of the D₂ reactant (270 ± 8 K) and the finite resolution of the TOF apparatus. A Monte Carlo simulation is carried out to account for these deviations. This simulation is calibrated using a measurement of the HD($v=4', J=3'$) product of the photolysis of HDS at 130.6 nm.³⁴ This analysis reveals an approximately Gaussian instrument function with a FWHM of $\Delta \cos \theta = 0.25$.

A modified singular-value-decomposition analysis of the Monte Carlo simulation is used to fit the experimental data (Fig. 2, thick line). The differential cross section is given in Fig. 3 where the scattering angle θ is measured from the incoming H atom to the outgoing HD product in the center-of-mass frame. Details of this analysis will be presented in a future work.³² We note, however, that the cross section presented in Fig. 3 is not the normalized differential cross section $(1/\sigma)(d\sigma/d\Omega)$, but rather the differential cross section blurred by a truncated Gaussian with a FWHM of $\Delta \cos \theta = 0.25$:

$$\frac{1}{\sigma} \frac{d\bar{\sigma}(x)}{d\Omega} = \frac{\int_{-1}^1 (1/\sigma) [d\sigma(x')/d\Omega] e^{-[(x-x')/\Delta x]^2} dx'}{\int_{-1}^1 e^{-[(x-x')/\Delta x]^2} dx'} \quad (3)$$

Here $x = \cos \theta$ and $\Delta x = \Delta \cos \theta$. We also note that to obtain the reported differential cross section we assume that the unblurred differential cross section may be approximated by a ninth-degree polynomial in $\cos \theta$.

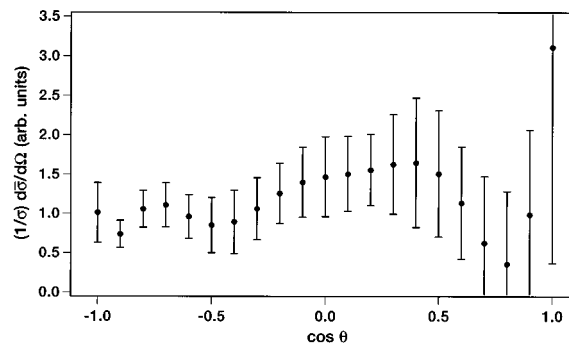


FIG. 3. State-specific differential cross section for the reaction $\text{H} + \text{D}_2 \rightarrow \text{HD} (v'=4, J'=3) + \text{D}$ at a collision energy of 2.2 eV. Error bars are dominated by the uncertainties in the characterization of the experimental apparatus. This differential cross section is recorded with a resolution of $\Delta \cos \theta = 0.25$.

The differential cross section reported here marks the first measurement of a rovibrational state-specific differential cross section of the $\text{H} + \text{H}_2$ reaction family in which the molecular product is detected directly at all scattering angles. Although the gross appearance of the differential cross section is flat (isotropic scattering), its profile suggests that two broad peaks centered around 75° and 140° exist in the differential cross section. These peaks can be better seen in the time-of-flight profiles, as indicated by the two arrows in Fig. 2. More quantitative statements about the structure of the cross section are presently limited by the large uncertainties, particularly in the forward region. These uncertainties are dominated by the precision of our instrument function. The resolution of this measurement is roughly 0.25 in $\cos \theta$. Neither the angular resolution nor the large uncertainties in the state-specific differential cross section are inherent limitations of the present experiment and are expected to be improved as the calibration of the instrument is refined.

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